## **AMENDMENTS TO THE CLAIMS**

This listing of claims will replace all prior versions, and listings, of claims in the application:

- 1. (Original) A catalyst system capable of catalysing the carbonylation of an ethylenically unsaturated compound, which system is obtainable by combining:
- a) a metal of Group VIB or Group VIIIB or a compound thereof,
- b) a bidentate phosphine, arsine, or stibine ligand, and
- c) an acid,

wherein said ligand is present in at least a 2:1 molar excess compared to said metal or said metal in said metal compound, and that said acid is present in at least a 2:1 molar excess compared to said ligand.

- 2. (Original) A catalyst system as claimed in claim 1 wherein the ratio of said ligand to said metal is in the range 5:1 to 750:1.
- 3. (Original) A catalyst system as claimed in claim 1 wherein the ratio of said ligand to said metal is in the range 10:1 to 500:1.
- 4. (Original) A catalyst system as claimed in claim 1 wherein the ratio of said ligand to said metal is in the range 20:1 to 40:1.
- 5. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any of the preceding elaims wherein the ratio of said acid to said ligand is in the range 5:1 to 95:1.

- 6. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any of the preceding elaims wherein the ratio of said acid to said ligand is in the range 20:1 to 40:1.
- (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any of the preceding elaims wherein the molar ratio of said acid to said metal is in the range 10:1 to 75000:1.
- 8. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any of the preceding elaims wherein the molar ratio of said acid to said metal is in the range 100:1 to 25000:1.
- 9. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any of the <u>preceding claims</u> wherein the molar ratio of said acid to said metal is in the range 200:1 to 400:1.
- 10. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any preceding <u>claim</u>-wherein said ligand is a bidentate phosphine ligand.
- 11. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any preceding claim wherein said ligand is of general formula (I)

(I)

wherein:

Ar is a bridging group comprising an optionally substituted aryl moiety to which the phosphorus atoms are linked on available adjacent carbon atoms;

A and B each independently represent lower alkylene;

K, D, E and Z are substituents of the aryl moiety (Ar) and each independently represent hydrogen, lower alkyl, aryl, Het, halo, cyano, nitro, OR<sup>19</sup>, OC(O)R<sup>20</sup>, C(O)R<sup>21</sup>, C(O)OR<sup>22</sup>, NR<sup>23</sup>R<sup>24</sup>, C(O)NR<sup>25</sup>R<sup>26</sup>, C(S)R<sup>25</sup>R<sup>26</sup>, SR<sup>27</sup>, C(O)SR<sup>27</sup>, or -J-

Q<sup>3</sup>(CR<sup>13</sup>(R<sup>14</sup>)(R<sup>15</sup>)CR<sup>16</sup>(R<sup>17</sup>)(R<sup>18</sup>) where J represents lower alkylene; or two adjacent groups selected from K, Z, D and E together with the carbon atoms of the aryl ring to which they are attached form a further phenyl ring, which is optionally substituted by one or more substituents selected from hydrogen, lower alkyl, halo, cyano, nitro, OR<sup>19</sup>, OC(O)R<sup>20</sup>, C(O)R<sup>21</sup>, C(O)OR<sup>22</sup>, NR<sup>23</sup>R<sup>24</sup>, C(O)NR<sup>25</sup>R<sup>26</sup>, C(S)R<sup>25</sup>R<sup>26</sup>, SR<sup>27</sup> or C(O)SR<sup>27</sup>;

 $R^{13}$  to  $R^{18}$  each independently represent lower alkyl, aryl, or Het;  $R^{19}$  to  $R^{27}$  each independently represent hydrogen, lower alkyl, aryl or Het;  $R^{1}$  to  $R^{12}$  each independently represent lower alkyl, aryl, or Het;  $Q^{1}$ ,  $Q^{2}$  and  $Q^{3}$  (when present) each independently represent phosphorous, arsenic or

antimony and in the latter two cases references to phosphine or phosphorous above are amended accordingly, with preferably both  $Q^1$  and  $Q^2$  representing phosphorus, more preferably all of  $Q^1$ ,  $Q^2$  and  $Q^3$  (when present) representing phosphorus.

12. (Original) A catalyst system as claimed in claim 11 wherein at least one (CR<sup>x</sup>R<sup>y</sup>R<sup>z</sup>) group attached to Q<sup>1</sup> and/or Q<sup>2</sup>, i.e. CR<sup>1</sup>R<sup>2</sup>R<sup>3</sup>, CR<sup>4</sup>R<sup>5</sup>R<sup>6</sup>, CR<sup>7</sup>R<sup>8</sup>R<sup>9</sup>, CR<sup>10</sup>R<sup>11</sup>R<sup>12</sup>, CR<sup>13</sup>R<sup>14</sup>R<sup>15</sup>, or CR<sup>16</sup>R<sup>17</sup>R<sup>18</sup>, may instead be represented by the group (Ad) wherein:

Ad each independently represent an optionally substituted adamantyl or congressyl radical bonded to the phosphorous atom via any one of its tertiary carbon atoms, the said optional substitution being by one or more substituents selected from hydrogen, lower alkyl, halo, cyano, nitro, OR<sup>19</sup>, OC(O)R<sup>20</sup>, C(O)R<sup>21</sup>, C(O)OR<sup>22</sup>,

NR<sup>23</sup>R<sup>24</sup>, C(O)NR<sup>25</sup> R<sup>26</sup>, C(S)R<sup>25</sup>R<sup>26</sup>, SR<sup>27</sup> or C(O)SR<sup>27</sup>; or if both (CR<sup>x</sup>R<sup>y</sup>R<sup>2</sup>) groups attached to either or both Q<sup>1</sup> and/or Q<sup>2</sup>, or Q<sup>3</sup> (if present) together with either Q<sup>1</sup> or Q<sup>2</sup> (or Q<sup>3</sup>) as appropriate, form an optionally substituted 2-phosphatricyclo[3.3.1.1{3,7}]decyl group or derivative thereof, or form a ring system of formula

wherein

 $R^{49}$ , and  $R^{54}$ , each independently represent hydrogen, lower alkyl or aryl;  $R^{50}$  to  $R^{53}$ , when present, each independently represent hydrogen, lower alkyl, aryl or Het; and

Y represents oxygen, sulfur or N-R<sup>55</sup>; and R<sup>55</sup>, when present, represents hydrogen, lower alkyl or aryl.

13. (Currently Amended) A catalyst system as claimed in claim 11 as claimed in either of claims 11 and 12 wherein said ligand is represented as:

$$(Ad)_{S}(CR^{7}R^{8}R^{9})_{T}Q^{2}-A-(K,D)Ar(E,Z)-B-Q^{1}(Ad)_{u}(CR^{1}R^{2}R^{3})_{v}\\$$

wherein Ar, A, B, K, D, E and Z,  $Q^1$ ,  $Q^2$ , and  $Q^3$ , and  $R^1$  to  $R^{27}$  are as defined in claim 11 except that K, D, E and Z may represent -J- $Q^3(Ad)_w(CR^{13}(R^{14})(R^{15})_x$  instead of -J- $Q^3(CR^{13}(R^{14})(R^{15}))CR^{16}(R^{17})(R^{18})$  and Ad is as defined in claim 12,

S & U = 0, 1 or 2 provided that 
$$S + U \ge 1$$
;  
T & V = 0, 1 or 2 provided that  $T + V \le 3$ ;

$$W \& X = 0, 1 \text{ or } 2.$$

14. (Currently Amended) A catalyst system as claimed in <u>claim 1</u> any of claims 1 to 10 wherein said ligand is of general formula (III):

$$X^4$$
 $Q^1-X^3$ 
 $D_1$ 
 $A_2$ 
 $X^2$ 
 $A_1$ 
 $(L_2)_m$ 
 $(L_1)_n$ 
 $(III)$ 

wherein:

A<sub>1</sub> and A<sub>2</sub>, and A<sub>3</sub>, A<sub>4</sub> and A<sub>5</sub> (when present), each independently represent lower alkylene;

 $K^1$  is selected from the group consisting of hydrogen, lower alkyl, aryl, Het, halo, cyano, nitro,  $-OR^{19}$ ,  $-OC(O)R^{20}$ ,  $-C(O)R^{21}$ ,  $-C(O)OR^{22}$ ,  $-N(R^{23})R^{24}$ ,  $-C(O)N(R^{25})R^{26}$ ,  $-C(S)(R^{27})R^{28}$ ,  $-SR^{29}$ ,  $-C(O)SR^{30}$ ,  $-CF_3$  or  $-A_3$ - $O^3(X^5)X^6$ ;

D<sup>1</sup> is selected from the group consisting of hydrogen, lower alkyl, aryl, Het, halo, cyano, nitro,  $-OR^{19}$ ,  $-OC(O)R^{20}$ ,  $-C(O)R^{21}$ ,  $-C(O)OR^{22}$ ,  $-N(R^{23})R^{24}$ , - $C(O)N(R^{25})R^{26}$ ,  $-C(S)(R^{27})R^{28}$ ,  $-SR^{29}$ ,  $-C(O)SR^{30}$ ,  $-CF_3$  or  $-A_4$ - $Q^4(X^7)X^8$ ;

E<sup>1</sup> is selected from the group consisting of hydrogen, lower alkyl, aryl, Het, halo,

cyano, nitro, 
$$-OR^{19}$$
,  $-OC(O)R^{20}$ ,  $-C(O)R^{21}$ ,  $-C(O)OR^{22}$ ,  $-N(R^{23})R^{24}$ ,  $-C(O)N(R^{25})R^{26}$ ,  $-C(S)(R^{27})R^{28}$ ,  $-SR^{29}$ ,  $-C(O)SR^{30}$ ,  $-CF_3$  or  $-A_5-Q^5(X^9)X^{10}$ ;

or both D<sup>1</sup> and E<sup>1</sup> together with the carbon atoms of the cyclopentadienyl ring to which they are attached form an optionally substituted phenyl ring:

 $X^1$  represents  $CR^1(R^2)(R^3)$ , congressyl or adamantyl,  $X^2$  represents  $CR^4(R^5)(R^6)$ , congressyl or adamantyl, or  $X^1$  and  $X^2$  together with  $Q^2$  to which they are attached form an optionally substituted 2-phospha-tricyclo[3.3.1.1{3,7}]decyl group or derivative thereof, or  $X^1$  and  $X^2$  together with  $Q^2$  to which they are attached form a ring system of formula IIIa

 $X^3$  represents  $CR^7(R^8)(R^9)$ , congressyl or adamantyl,  $X^4$  represents  $CR^{10}(R^{11})(R^{12})$ , congressyl or adamantyl, or  $X^3$  and  $X^4$  together with  $Q^1$  to which they are attached form an optionally substituted 2-phospha-tricyclo[3.3.1.1{3,7}]decyl group or derivative thereof, or  $X^3$  and  $X^4$  together with  $Q^1$  to which they are attached form a ring system of formula IIIb

X<sup>5</sup> represents CR<sup>13</sup>(R<sup>14</sup>)(R<sup>15</sup>), congressyl or adamantyl, X<sup>6</sup> represents

 $CR^{16}(R^{17})(R^{18})$ , congressyl or adamantyl, or  $X^5$  and  $X^6$  together with  $Q^3$  to which they are attached form an optionally substituted 2-phosphatricyclo[3.3.1.1{3,7}]decyl group or derivative thereof, or  $X^5$  and  $X^6$  together with  $Q^3$  to which they are attached form a ring system of formula IIIc

 $X^7$  represents  $CR^{31}(R^{32})(R^{33})$ , congressyl or adamantyl,  $X^8$  represents  $CR^{34}(R^{35})(R^{36})$ , congressyl or adamantyl, or  $X^7$  and  $X^8$  together with  $Q^4$  to which they are attached form an optionally substituted 2-phosphatricyclo[3.3.1.1{3,7}]decyl group or derivative thereof, or  $X^7$  and  $X^8$  together with  $Q^4$  to which they are attached form a ring system of formula IIId

 $X^9$  represents  $CR^{37}(R^{38})(R^{39})$ , congressyl or adamantyl,  $X^{10}$  represents  $CR^{40}(R^{41})(R^{42})$ , congressyl or adamantyl, or  $X^9$  and  $X^{10}$  together with  $Q^5$  to which they are attached form an optionally substituted 2-phosphatricyclo[3.3.1.1.{3,7}]decyl group or derivative thereof, or  $X^9$  and  $X^{10}$  together with  $Q^5$  to which they are attached form a ring system of formula IIIe

 $Q^1$  and  $Q^2$ , and  $Q^3$ ,  $Q^4$  and  $Q^5$  (when present), each independently represent

phosphorus, arsenic or antimony;

M represents a Group VIB or VIIIB metal or metal cation thereof;

L<sub>1</sub> represents an optionally substituted cyclopentadienyl, indenyl or aryl group;

L<sub>2</sub> represents one or more ligands each of which are independently selected from hydrogen, lower alkyl, alkylaryl, halo, CO, P(R<sup>43</sup>)(R<sup>44</sup>)R<sup>45</sup> or N(R<sup>46</sup>)(R<sup>47</sup>)R<sup>48</sup>;

R<sup>1</sup> to R<sup>18</sup> and R<sup>31</sup> to R<sup>42</sup>, when present, each independently represent hydrogen, lower alkyl, aryl, halo or Het;

R<sup>19</sup> to R<sup>30</sup> and R<sup>43</sup> to R<sup>48</sup>, when present, each independently represent hydrogen, lower alkyl, aryl or Het;

R<sup>49</sup>, R<sup>54</sup> and R<sup>55</sup>, when present, each independently represent hydrogen, lower alkyl or aryl;

R<sup>50</sup> to R<sup>53</sup>, when present, each independently represent hydrogen, lower alkyl, aryl or Het;

Y<sup>1</sup>, Y<sup>2</sup>, Y<sup>3</sup>, Y<sup>4</sup> and Y<sup>5</sup>, when present, each independently represent oxygen, sulfur or N-R<sup>55</sup>;

$$n = 0 \text{ or } 1;$$

and m = 0 to 5;

provided that when n = 1 then m equals 0, and when n equals 0 then m does not equal 0.

- 15. (Original) A catalyst system as claimed in claim 14 wherein if both  $K^1$  represents  $A_3$ - $Q^3(X^5)X^6$  and  $E^1$  represents - $A_5$ - $Q^5(X^9)X^{10}$ , then  $D^1$  represents - $A_4$ - $Q^4(X^7)X^8$ .
- 16. (Currently Amended) A catalyst system as claimed in <u>claim 12</u> any of claims 12 to 15, wherein adamantyl represents unsubstituted adamantyl or adamantyl substituted with one or more unsubstituted C<sub>1</sub>-C<sub>8</sub> alkyl substituents, or a combination thereof.
- 17. (Currently Amended) A catalyst system as claimed in <u>claim 12</u> any of claims 12 to 16, wherein 2-phospha-adamantyl represents unsubstituted 2-phospha-adamantyl or 2-phospha-adamantyl substituted with one or more unsubstituted C<sub>1</sub>-C<sub>8</sub> alkyl substituents, or a combination thereof.
- 18. (Currently Amended) A catalyst system as claimed in <u>claim 12</u> any of claims 12 to 17, wherein 2-phospha-adamantyl includes one or more oxygen atoms in the 2-phospha-adamantyl skeleton.
- 19. (Currently Amended) A catalyst system as claimed in <u>claim 12</u> any of claims 12 to
   18, wherein congressyl represents unsubstituted congressyl.

- 20. (Currently Amended) A catalyst system according to <u>claim 1</u> any preceding claim wherein the metal or compound thereof is palladium.
- 21. (Original) A catalyst system according to claim 20 wherein the palladium is in the metal form.
- 22. (Currently Amended) A catalyst system according to <u>claim 1</u> any preceding claim, wherein the catalyst system includes in a liquid reaction medium a polymeric dispersant dissolved in a liquid carrier, said polymeric dispersant being capable of stabilising a colloidal suspension of particles of the Group VI or VIIIB metal or metal compound of the catalyst system within the liquid carrier.
  - 23. (Currently Amended) A process for the carbonylation of an ethylenically unsaturated compound comprising contacting an ethylenically unsaturated compound with carbon monoxide and a hydroxyl group containing compound in the presence of a catalyst system as claimed in claim 1 any preceding claim.
  - 24. (Original) A process according to claim 23, wherein the carbonylation of an ethylenically unsaturated compound is performed in one or more aprotic solvents.
  - 25. (Currently Amended) A process according to claim 23 either of claims 23 and 24, wherein suitable ethylenically unsaturated compounds include ethene, propene, hexene, vinyl compounds such as vinyl acetates, heptene, octene, nonene, decene, undecene, dodecene, etc up to C<sub>30</sub>, i.e. having from 2 to 30 carbon atoms, which may be linear or branched, cyclic or uncyclic or part cyclic and in which the double bond may take any suitable position in the carbon chain and which includes all stereisomers thereof.

26. (Original) A reaction medium comprising one or more reactants, and a catalyst system comprising, or obtainable by combining, at least a Group VIB or VIIIB metal or metal compound, a bidentate phosphine, arsine, or stibine ligand, and an acid, wherein said ligand is present in at least a 2:1 molar excess compared to said metal or said metal in said metal compound, and that said acid is present in at least a 2:1 molar excess compared to said ligand.

- 27. (Currently Amended) A reaction medium comprising one or more reactants, and a catalyst system as claimed in claim 1 any one of claims 1 to 22.
- 28. (Currently Amended) A reaction medium as claimed in <u>claim 26</u> either of claims 26 and 27 wherein the amount of free acid present in the medium is greater than 500ppm.
- 29. (Original) Use of a system comprising, or obtainable by combining:
- a) a metal of Group VIB or Group VIIIB or a compound thereof,
- b) a bidentate phosphine, arsine, or stibine ligand, preferably a bidentate phosphine ligand, and
- c) an acid,

wherein said ligand is present in at least a 2:1 molar excess compared to said metal or said metal in said metal compound, and that said acid is present in at least a 2:1 molar excess compared to said ligand, as a catalyst in the carbonylation of an ethylenically unsaturated compound.

- 30. (Cancelled)
- 31. (Cancelled)

- 32. (Cancelled)
- 33. (Cancelled)
- 34. (Original) A catalyst system capable of catalysing the carbonylation of an ethylenically unsaturated compound, said system comprising:
- a) a metal of Group VIB or Group VIIIB or a compound thereof,
- b) a bidentate phosphine, arsine, or stibine ligand, preferably a bidentate phosphine ligand, and
- c) an acid,

wherein said ligand is present in at least a 2:1 molar excess compared to said metal or said metal in said metal compound, and that said acid is present in at least a 2:1 molar excess compared to said ligand.

- 35. (Original) A process for the carbonylation of an ethylenically unsaturated compound comprising contacting an ethylenically unsaturated compound with carbon monoxide and a hydroxyl group containing compound in the presence of a catalyst system, said system comprising:
- a) a metal of Group VIB or Group VIIIB or a compound thereof,

molar excess compared to said ligand.

- b) a bidentate phosphine, arsine, or stibine ligand, preferably a bidentate phosphine ligand, and
- an acid,
   wherein said ligand is present in at least a 2:1 molar excess compared to said metal
   or said metal in said metal compound, and that said acid is present in at least a 2:1

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36. (Original) A complex capable of catalysing the carbonylation of an ethylenically unsaturated compound, said complex obtainable by combining:

- a) a metal of Group VIB or Group VIIIB or a compound thereof,
- b) a bidentate phosphine, arsine, or stibine ligand, preferably a bidentate phosphine ligand, and
- c) an acid,

wherein said ligand is present in at least a 2:1 molar excess compared to said metal or said metal in said metal compound, and that said acid is present in at least a 2:1 molar excess compared to said ligand.

37. (Original) A process for the carbonylation of an ethylenically unsaturated compound with carbon monoxide and a hydroxyl group containing compound in the presence of a complex, said complex as defined in claim 36.